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M. J. Murphy

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# An Improved Reaction Rate Equation for Simulating the Ignition and Growth of Reaction in High Explosives

Michael J. Murphy  
Lawrence Livermore National Laboratory  
P.O. Box 808, L-099, Livermore, CA 94550 USA

**Abstract.** We describe an improved reaction rate equation for simulating ignition and growth of reaction in high explosives. It has been implemented into CALE<sup>1</sup> and ALE3D<sup>2</sup> as an alternate to the baseline the Lee-Tarver reactive flow model<sup>3,4</sup>. The reactive flow model treats the explosive in two phases (unreacted/reactants and reacted/products) with a reaction rate equation to determine the fraction reacted,  $F$ . The improved rate equation has fewer parameters, is continuous with continuous derivative, results in a unique set of reaction rate parameters for each explosive while providing the same functionality as the baseline rate equation. The improved rate equation uses a cosine function in the ignition term and a sine function in the growth and completion terms. The improved rate equation is simpler with fewer parameters

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## Introduction

The reactive flow (RF) model for simulating shock initiation of energetic materials has been in use since the early 1980's<sup>3</sup>. The model is based on an ignition and growth of reaction rate equation for modeling high explosives as they transition from the unreacted state to the reacted state. A JWL form of equation of state (EOS) is used to describe the response of the reacted (explosive products). An equation of state, typically JWL form, is also used to describe the response of the unreacted (inert) material. The reaction rate equation is used to determine the amount of the material that reacts every cycle (fraction reacted) and the transition from the unreacted to the reacted state.

In this paper we describe a modified form of the ignition and growth of reaction rate equation (aka reactive flow, RF) that uses a half period of the cosine function in the ignition term and a half period of the sine function in the growth of reaction and completion terms. The improved rate equation is simpler with fewer parameters while

maintaining the functionality of the baseline rate equation. It is better suited for optimization because the parameters are not correlated and the maximum value of the growth and completion term form factor (FF) is always 1.0.

## Baseline Rate Equation

There are two versions of the baseline RF rate equation that have been published. The two-term form, Eq. 1, has an ignition term and a single growth of reaction term<sup>4</sup>. It has been shown to be useful for a wide range of shock and impact initiation studies. The three-term form of the rate equation, Eq. 2, consists of an ignition term, a growth of reaction term, and a completion term<sup>5</sup>. The three-term form has been shown to be more effective than the two-term form for high intensity, short duration shocks. However, for initiation problems that do not involve high pressure short pulse duration shocks, the two-term form of the rate equation can be successfully used while also having less parameters<sup>6</sup>.

$$dF/dt = Freq \cdot (1-F)^{Frer} \cdot (\rho/\rho_0 - 1 - Ccrit)^{Eeta1} + Grow \cdot (1-F)^{Es} \cdot F^{Ar} \cdot p^{em} \quad (1)$$

$$dF/dt = Freq \cdot (1-F)^{Frer} \cdot (\rho/\rho_0 - 1 - Ccrit)^{Eeta1} + Grow_1 \cdot (1-F)^{Es1} \cdot F^{Ar1} \cdot p^{em} + Grow_2 \cdot (1-F)^{Es2} \cdot F^{Ar2} \cdot p^{en} \quad (2)$$

$$dF/dt = Freq \cdot 0.5 \cdot (1 + \cos(\pi F / figmx)) (\rho/\rho_0 - 1 - Ccrit)^{Eeta1} + Grow_2 \cdot \sin(\pi F^{Es}) \cdot p^{en} \quad (3)$$

$$dF/dt = Freq \cdot 0.5 \cdot (1 + \cos(\pi F / figmx)) (\rho/\rho_0 - 1 - Ccrit)^{Eeta1} + Grow_1 \cdot \sin(\pi F^{Es1}) \cdot p^{em} + Grow_2 \cdot \sin(\pi F^{Es}) \cdot p^{en} \quad (4)$$

The utility of the baseline forms of the rate equation (Eq. 1 & 2) have been demonstrated for many materials. However, it is difficult to determine optimal parameter sets because:

- The ignition term causes a discontinuity in  $df/dt$  when  $F$  exceeds the ignition limit.
- The maximum values of the growth and completion term FFs vary as the  $E_s$  and  $A_r$  parameter values are varied.
- The  $Grow$ ,  $E_s$ , &  $A_r$  variables are correlated making it difficult to parameterize and optimize.

### New Rate Equation

The two-term and three-term forms of the new rate equation are described in Eq. 3 & 4. The three-term forms of the baseline and new rate equations (Eq 2 & 4) default to the two-term forms (Eq. 1 & 3) when the  $Grow_1$  term is set equal to zero. The new form of the reaction rate equation has several advantages for optimization over the baseline form:

- The ignition term is continuous and smoothly goes to 0.0 as  $F$  goes to the ignition limit ( $figmx$ ).
- The maximum value of the growth and completion FFs is always 1.0.
- The  $Grow$  &  $E_s$  variables are not correlated making it easy to parameterize and optimize.
- There are fewer parameters.

### Comparison of Ignition Terms

A graphical comparison of the ignition term portion of the baseline and new rate equations is shown in Fig. 1. The red curve shows a plot of the ignition portion of the baseline rate equation as a function of the fraction reacted. It shows how the ignition term is discontinuous when the fraction reacted ( $F$ ) reaches the ignition limit,  $figmx$ . The new ignition term, shown in blue, is continuous and smoothly approaches a value of 0.0 as  $F$  approaches the ignition limit.

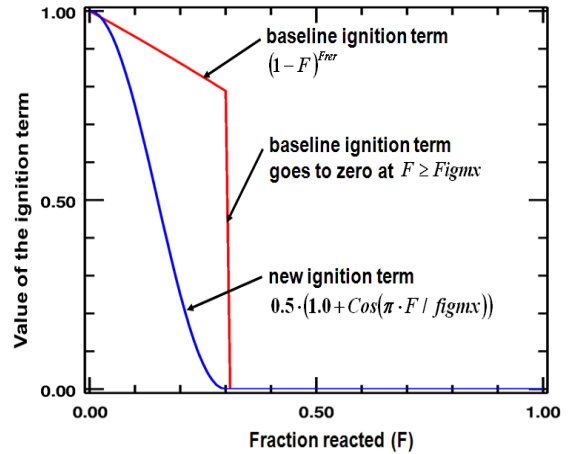


Fig. 1. Comparison of baseline and improved rate equation ignition terms

### Comparison of Growth Terms

A comparison of the FFs for the baseline and new rate equations is discussed in the following section. This discussion applies to either the growth or completion terms of the rate equation.

#### Baseline Growth/Completion Term Form Factor

The FF given in Eq. 5 is used in the growth & completion terms of the baseline rate equation. Three example curves showing the value of this FF as a function of the fraction reacted,  $F$ , are shown in Fig. 2. The curves are for three sets of values of  $E_s$  and  $A_r$ . The maximum value of the FF is different for each set of  $E_s$  and  $A_r$  values. Changes in  $E_s$  or  $A_r$  in order to affect a shift in the shape of the FF curve (i.e. fast rise vs. slow rise) may also a result in a change in the peak value of the curve.

$$(1-F)^{Es} \cdot F^{Ar} \quad (5)$$

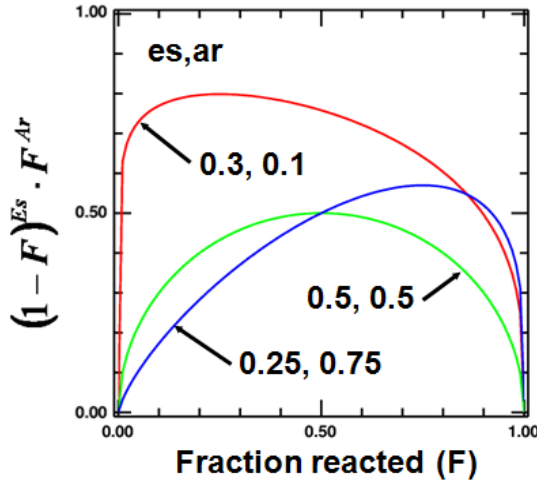


Fig. 2. Three curves of the baseline FF as a function of the fraction reacted.

#### New Growth Term Form Factor

The new FF for the growth and completion terms of the rate equation is based on a half period of the sine function as shown in Eq. 6.

$$\text{Sin}(\pi F^{E_s}) \quad (6)$$

The new FF has one less parameter than in the baseline equation and always has a maximum value of 1.0. Three curves showing the value of the new FF for three values of  $E_s$  are plotted as a function of the fraction reacted,  $F$ , in Fig. 3. The sine function starts at zero with  $F$  equal to zero (unreacted). It reaches a maximum of 1.0 when  $F^{E_s}$  is equal to 0.5 and goes back to zero as  $F$  goes to 1.0 (fully reacted). The parameterization process is simplified because  $E_s$  can be altered to shift the curve in  $F$  space (fast rise vs. slow rise) without changing the maximum value of the FF. The sine function normalizes the FF and we expect that the value of the growth and completion prefactor terms ( $Grow_1$  &  $Grow_2$ ) should be correlated to the shock to detonation transition characteristics of the explosive. Additional benefits of the new FF are that the derivatives are continuous. It may also be possible to not use the ending and starting limits on the growth and completion terms of the three-term model by making  $E_s$  small or large.

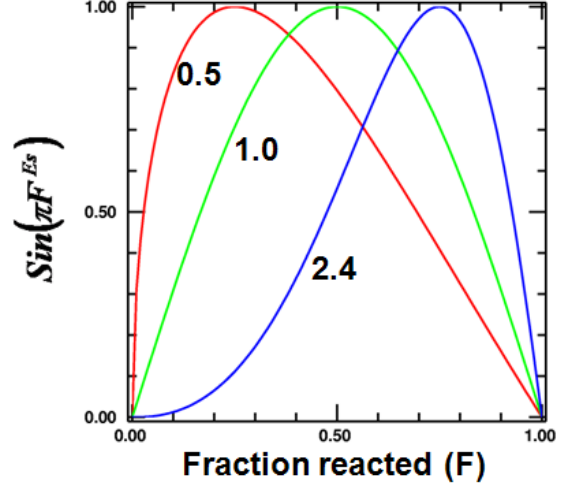


Fig. 3. Three curves of the new FF as a function of the fraction reacted.

#### Parameter Optimization

We have made a comparison of parameter optimization for the baseline rate equation with parameter optimization for the new rate equation. The parameter values were determined using GLO<sup>5</sup> by simulating the results of two EDC-37 shock-to-detonation (SDT) embedded particle velocity gage experiments<sup>6</sup>. The simulations were run with ALE3D and CALE. The GLO determined baseline RF initiation parameters were verified by comparing to the results of two additional SDT embedded gage experiments. The first part of the parameterization process involved determining the unreacted equation of state of the EDC-37.

#### Unreacted Equation of State

We determined the unreacted EOS parameters by simulating SDT experiments with the reaction rate parameters set to zero and matching to the early portion of the experimental data. The Gruneisen EOS parameters ( $c_0$  &  $s_1$ ) were first determined. The JWL EOS parameters values ( $a$ ,  $b$ ,  $r1$ , &  $r2$ ) for the unreacted material were then determined directly from the Gruneisen EOS parameter values by finding the best fit of the JWL pressure-volume curve to the Gruneisen pressure-volume curve in the relative volume range of 0.75 to 1.0.

## Baseline RF Rate Equation Parameters

The optimum parameter values for the baseline two-term form reaction rate equation were found with a combination of global & local optimization. Five hundred global values of  $A_r$  and  $E_s$  were first defined using latin hypercube sampling<sup>9</sup>. For each value of  $A_r$  &  $E_s$ , the values of  $Freq$  and  $Grow$  were locally optimized to find the best match to the experimental data as defined by a figure of merit (FOM). Plots of the locally optimized value of the FOM versus the globally defined values of  $A_r$  and  $E_s$  are shown below in Figs 4 & 5.

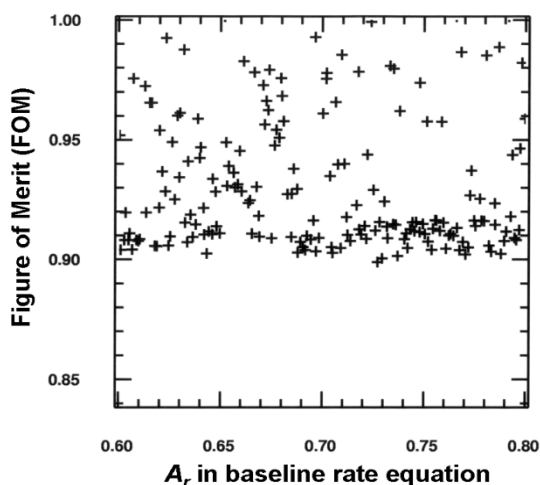


Fig. 4. Optimized FOM vs  $A_r$  with the baseline Eq.

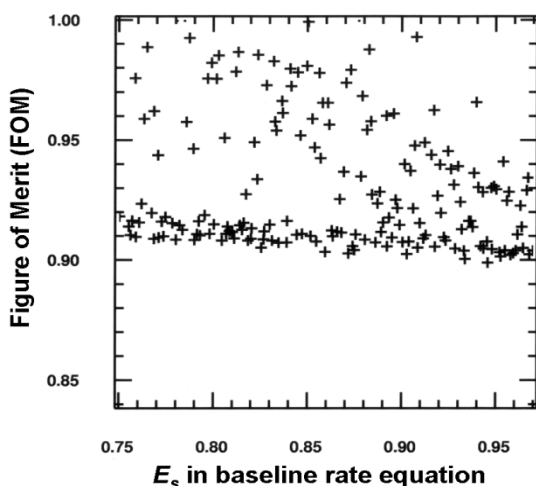


Fig. 5. Optimized FOM vs  $E_s$  with the baseline Eq.

A plot of the FOM versus the locally optimized values of  $Grow$  is shown in Fig 6. Figures 4, 5, & 6 show the difficulty of determining an optimum set of values for the parameters used in the baseline rate equation. There are many good values but there is not a “best” set of values. The plots show a slight improvement in the FOM with increasing values of  $E_s$  and  $Grow$ . However, a subsequent study with larger values of  $E_s$  continued with the same trend without resulting in an obvious set of optimum parameter values.

We also tried normalizing the growth of reaction term by dividing  $Grow$  by the maximum value of the FF (Eq. 5) for each set of  $A_r$  and  $E_s$ . This improved the optimization process, however, it still did not lead to an obvious best set of parameter values. The bottom line is that the three parameters used in the growth of reaction term are primarily used to define two things; 1) the values of the fraction reacted,  $F$ , where the reaction rate term is at a maximum, and 2) the maximum value of the reaction rate term.

Our conclusion is that the parameters used in the baseline reaction rate equation result in an “over specified” condition which can lead to multiple sets of values that give good results. This makes it difficult to optimize because widely varying parameter values can give about the same result.

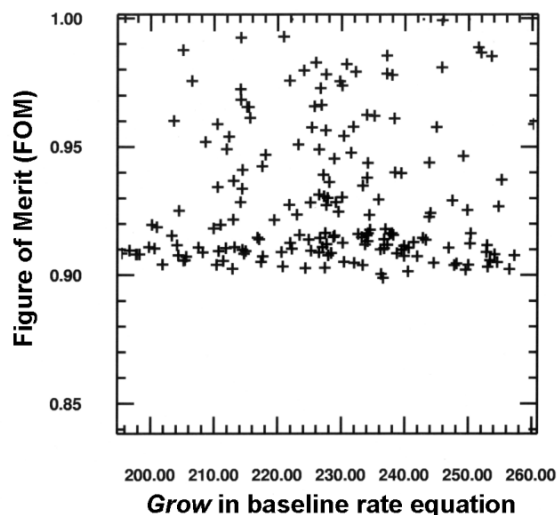


Fig.6. Optimized FOM vs  $Grow$  with baseline Eq.

## New Rate Equation Parameters

The optimum parameter values for the new two-term form reaction rate equation were found in the same way as with the baseline. We used a combination of global & local optimization. Five hundred global values of  $E_s$  (Ar is not required) were first defined using latin hypercube sampling. For each value of  $E_s$ , the ignition term,  $Freq$ , and the growth of reaction term,  $Grow$  were locally optimized to find the best match to the experimental data. A plot of the locally optimized value of the FOM versus the globally defined values of  $E_s$  is shown below in Fig. 7.

This plot has a nice parabolic shape with a minimum at about  $E_s = 0.81$ . It also shows the utility of using the sine function in the FF, as there is a fairly obvious optimum value for  $E_s$ .

A plot of the FOM as a function of the locally optimized value of the growth of reaction parameter ( $Grow$ ) is shown in Fig. 8. This plot also has a nice parabolic shape with a minimum at about  $Grow = 85$ . A similar result was obtained for the value of  $Freq$ .

The global and local optimization studies show that the parameters in the new form of the reaction rate equation are not correlated and have uniquely optimum values. In addition, the new rate equation has fewer parameters and a lower FOM, 0.85 versus 0.92, showing a better match to experiment than the baseline rate equation

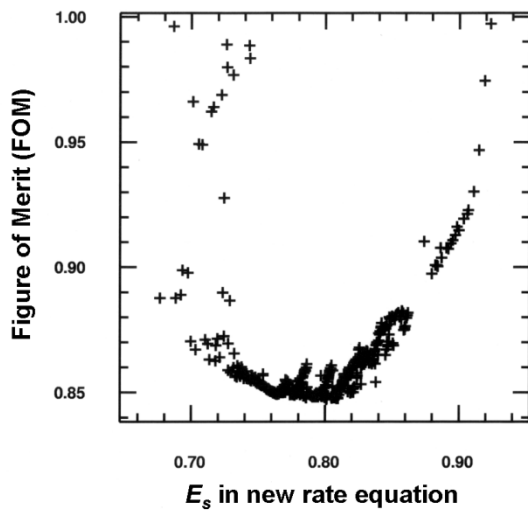


Fig. 7. Optimized FOM vs  $E_s$  with the new Eq.

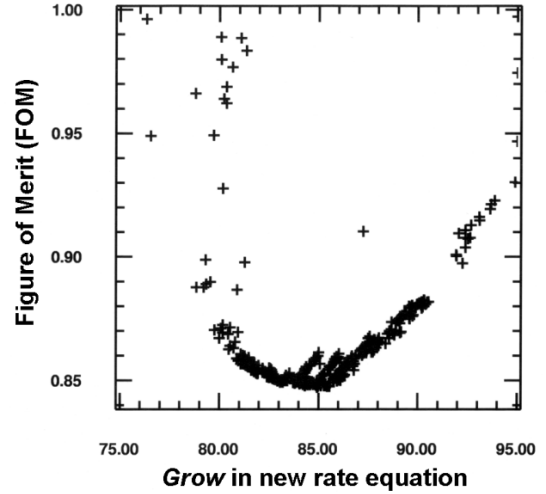


Fig. 8. Optimized FOM vs  $Grow$  with the new Eq.

We observe that the best set of parameter values for the baseline equation ( $A_r=0.73$ ,  $E_s=0.95$ , and  $Grow=236$ ) result in a maximum in the FF at  $F=0.435$ . Similarly, for a value of 0777 for  $E_s$  in the new rate equation results in a maximum in the FF at  $F=0.412$ . Looking at the growth of reaction term we see a very comparable result. The maximum value of the baseline equation FF (Eq. 5, at  $A_r=0.73$ , and  $E_s=0.95$ ) is 0.3166. Multiplying this by the optimal baseline  $Grow$  value, 236, yields a normalized value of 74.7 which is quite close to the optimal  $Grow$  value of 83.6 that was obtained for the new rate equation.

## Parameters Values for EDC37

The optimized parameter values for EDC37 using the baseline and new rate equations are compared in Table 1. Note that some values have up to 7 significant digits – this is used to specify specific sets of parameters. The following key points can be observed from the comparison:

- The  $frer$  term is not used in the new rate Eq.
- The  $ilim$  term is 0.2 in the new rate Eq.
- The EDC37 parameter set uses the two term form of the rate equation ( $grow1$  terms not used).
- The  $Ar_2$  term is not used in the new rate Eq.
- The value of  $F$  at FF max for the baseline and new Eqs are 0.435 and 0.412 respectively.
- The normalized  $Grow$  (74.7) for the baseline Eq. is about the same as  $Grow$  (83.6) with the new Eq.

## Parameters Values for CompB

The parameter values for CompB from Reference 6 are listed in Table 1 under the “Old Baseline CompB” column. These values were not “optimized” with GLO at the time of parameterization. The optimized values for the new rate equation are listed in the table under the “New Eq. CompB” column.

A conversion of parameters back to the baseline model from the optimized “New Eq.” parameter set is listed in the “new Baseline CompB” column. The following steps were followed in the conversion:

- Set  $Es_2=0.88$ .
- Set  $Ar_2=0.734$  so that  $FF_{\max}$  occurs at  $F=0.455$
- Determine optimal  $Grow=681$ .

## Conclusions

Fitting the coefficients of the unreacted JWL by matching to a calibrated Gruneisen EOS is a simple and accurate approach. The combination of global and local optimization provides an effective way to determine optimized reaction rate coefficients from multiple experiments. The new reaction rate equation has several advantages over the baseline reaction rate equation:

- The ignition term is continuous and the derivatives are continuous.
- The maximum value of the growth and completion FF terms are always 1.0.
- The  $Grow$  &  $E_s$  parameters are not correlated making it easy to parameterize and optimize.
- The new rate equation has fewer parameters, and provides the same functionality as the baseline reaction rate equation.

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Table 1. Parameter values for the optimized baseline two-term RF model and for the improved two-term RF model for EDC37 and CompB

Parameter name	Baseline EDC37	New Eq. EDC37	Old Baseline CompB	New Eq. CompB	New Baseline CompB
refdp	1.842	1.842	1.630	1.630	1.630
ap	6.4051948	6.4051948	5.57483	5.57483	5.57483
bp	0.2510169	0.2510169	0.078301	0.078301	0.078301
rp1	4.25	4.25	4.50	4.50	4.50
rp2	1.825	1.825	1.20	1.20	1.20
wp	0.25	0.25	0.34	0.34	0.34
cmp	1.0e-05	1.0e-05	1.0e-05	1.0e-05	1.0e-05
q	0.134712	0.1347127	0.081	0.081	0.081
refdr	1.842	1.842	1.630	1.630	1.630
ar	12487	12487	1479	405.6	405.6
br	-1.7359e-4	-1.7359e-4	-5.261e-2	-2.991814e-3	-2.991814e-3
rr1	14.99	14.99	12.0	10.63	10.63
rr2	-4.07944	-4.07944	1.20	-1.71146	-1.71146
wr	0.8578	0.8578	0.9120	0.9120	0.9120
cmr	2.505e-5	2.505e-5	2.4868e-5	2.4868e-5	2.4868e-5
freq	2.524518e+1	3.738198e+1	44.0	44.0	44.0
frer	0.222	----	0.222	---	0.222
ccrit	0.0	0.0	0.01	0.01	0.01
eeta1	4.0	4.0	4.0	4.0	4.0
ilim	0.3	0.2	0.3	0.3	0.3
grow1	----	----	----	----	----
es1	----	----	----	----	----
ar1	----	----	----	----	----
em	----	----	----	----	----
glim	----	----	----	----	----
grow2	236	8.361347e+1	514.0	353.09	680
es2	0.95	0.7774716	0.222	0.88	0.88
ar2	0.73	----	0.667	----	0.734
en	2.0	2.0	2.0	2.0	2.0
clim	0.0	0.0	0.0	0.0	0.0
eps	0.001	0.001	0.001	0.001	0.001
t0	298.0	298.0	298.0	298.0	298.0
fcut	1.0e-10	1.0e-10	1.0e-10	1.0e-10	1.0e-10
brnli	0.03	0.03	0.03	0.03	0.03
beta	0.0	0.0	0.0	0.0	0.0
F @ FF max	0.435	0.412	0.750	0.455	0.455
FF @ FF max	0.3166	1.0	0.6067	1.0	0.333
G * FF @ FF max	74.7	83.6	311.8	353.09	226.4